

REFERENCES

1. Klimisch, R.L., and Komarmy, J.M. In R.L. Klimisch and L.G. Larson (eds.), The Catalytic Chemistry of Nitrogen Oxides, 305.
2. Iwamoto, M., and Hamada, H., Catal. Today, 10 , pp. 57, 1991.
3. Li, Y., and Hall, K. H., J. Catal., 129, pp. 202, 1991.
4. Iwamoto, M., Yahiro, H., Tanda, K., Mine, Y., and Kagawa, S., Phys. Chem., 95, 37275, 1995.
5. Hightower, J.W., and Von Leirsberg, D.A. In R.L. Klimisch and L.G. Larson (eds.), The Catalytic Chemistry of Nitrogen Oxides, 63.
6. Nakatsuji, T., and Miyamoto, A., Catal. Today, 10, pp. 21-31, 1991.
7. Cohn, G., Steek, D., and Andersen, H. 1961 U.S. Patent 2,975,025.
8. Bosch, H., and Janssen, F., Catal. Today, 2, pp. 369, 1988.
9. Hamada, H., Kintaichi, Y., Sasaki, M., Ito, T., and Tabata, M., Appl. Catal. , 64, pp. L1-L4, 1990.
10. Ukisu, Y., Sato, S., Muramatsu, G., and Yoshida, K. , Catal. Lett. , 16 pp. 11-16, 1992.
11. Iwamoto, M., Yahiro, H., Yoshiota, T., and Mizuno, N., Chem. Lett., 11 pp. 1967, 1990.
12. Inui, T., Kojo, S., Shibata, M., Yoshida, T., and Iwamoto, S., Stud. Surf. Sci. Catal., 69, pp. 355, 1991.
13. Kikuchi, E., Yogo, K., Tanaka, S., and Abe, M., Chem. Lett., pp. 1063, 1991.
14. Held, W., and Koenig, A., Richter, T., and Puppe, L., SAE paper, 900496 , 1990.

15. Iwamoto, M., Proceedings of Meeting of Catalytic Technology for Removal of Nitrogen Oxides, January 25, 1990, Tokyo, Japan, Catal. Soc. of Japan, 17, 1990.
16. Iwamoto, M., Yahiro, H., Shundo, S., Yu-u, Y. and Mizuno, N., Shokubai, 33, pp. 430, 1990.
17. Yogo, K., Ihara, M., Terasaki, I., and Kikuchi, E., Appl. Catal. B, 2, pp. L1-L5, 1993.
18. Bennett, C.J., Bennett, P.S., Golunski, S.E., Hayes, J.W., and Walker, A.P., Appl. Catal., 86, pp. L1-L6, 1992.
19. Petunchi, J.O., and Hall, W.K., Appl. Catal. B, 3, pp. 239-257, 1994.
20. Teraoka, Y., Pgawa, H., Furukawa, H., and Kagawa, S., Catal. Lett., 12, pp. 361-388, 1992.
20. Montreuil, C.N., and Shelef, M., Appl. Catal. B, 1, pp. L1-L8, 1992.
21. Lee, C.Y., Choi, K.Y., and Ha, B.H., Appl. Catal. B, 5, pp. 7-21, 1994.
22. Hamada, H. , Kintaichi, Y., Sasaki, M., and Ito, T., Chem. Lett., pp. 1069, 1990.
23. Iwamoto, M., Yahiro, H., and Tanda, K., Sucessful Design of Catalysis, pp. 219-226, Elsevier, Amsterdam, 1988.
24. Iwamoto, M., Sato, S., Yu-u, H. Yahiro, H., and Mizuno, N., Appl. Catal., 70, pp. L1-L5, 1991.
25. Iwamoto, M., Mizuno, N., and Yahiro, H., Sekiyu Gakkaishi, 34, pp. 375-390, 1991.
26. Iwamoto, M. and Mizuno, N., Proc. Instn. Mech. Engrs., 207, pp. 23-33, 1993.

27. Hamada, H., Kintaichi, K., Sasaki, M., Ito, T., and Tanabe, M., Appl. Catal., 70, pp. L15-L20, 1991.
28. Campa, M.C., Catal. Lett., 23, pp. 141-149, 1994.
29. Iwamoto, M., Konno, M., Chidahisa, T., and Murayama, SAE Paper, 920091, pp. 1-8, 1992.
30. Teraoka, Y., Catal. Lett., 12, pp. 361-366, 1992.
31. Yang, R.T., and Li, W., J. of Catal., 155, pp. 414-417, 1995.
32. Inui, T., Hirabayashi, T., and Iwamoto, S., Catal. Lett., 27, pp. 267-272, 1994.
33. Zhang, Y., Catal. Lett., 31, pp. L75-89, 1995.
34. Inui, T., Iwamoto, S., Kojo, S., and Yoshida, T., Catal. Lett., 16, pp. 223-229, 1992.
35. Li, Y., and Armor, J. N., Appl. Catal. B. Env., 5, pp. L257-L270, 1995.
36. Iwamoto, M., Mizuno, N. and Yahiro, H., Proc. 10 th Int. Cong. Catal., pp. 213-215, Budapest, 1992.
37. J. Pincharoenthaworn. Effect of of Hydrocarbons on the Performance of Cu/Na-MFI Catalyst for NO Removal. Master 's Thesis, Chulalongkorn University, 1995.
38. Flanigen, E.M. Zeolites and molecular sieves : an histrorical perspective. In H.V. Bekkum (ed.), Stud. Sur. Sci. Catal., 58 : Introduction to Zeolite Science and Practice, pp. 13-34. Netherlands, Elsevier, 1991.
39. Breck, D.W., Zeolite Molecular Sieves, New York, Robert E. Krieger Publishing Co., 1984.

40. Szostak, R. Molecular Sieves : Principles of Synthesis and Identification, New York, Van Nostrand Reinhold, 1989.
41. Khouw, C.B., and Davis, M.E., J. Am. Chem. Soc., 115, pp. 207-221, 1993.
42. Tanabe, K., Misono, M., Ona, Y., and Hattori, H. Acid and base centers : structure and acid-base property. In B. Delman and J.T. Yates (eds.), Stud. Sur. Sci. Catal., 51 : New Solid Acids and Bases, pp. 142-161, Tokyo, Elsevier, 1989.
43. Knouw, C. B., and Davis, M. E. Shape-Selectivity Catalysts with Zeolites and Molecular Sieve. In M.E. Davis and S.L. Suib (eds.), ACS Symposium Series 517 : Selectivity in Catalysis, pp. 206-211,, Washington , 1993.
44. Meier, W.M., and Olson, D.H., Atlas of Zeolite structure Types, Boston : Butterworths , 1987.
45. Gates, B.C., Catalytic Chemistry, Singapore, John Wiley & Sons, 1992.
46. Kokotailo G.T. Zeolite crystallography. In F.R. Ribeiro et al. (eds.), Zeolites : Science and Technology, pp. 83-108, Netherlands, Martinus Nijhoff Publishers, 1984.
47. Barthomeuf, D. Acidic catalysis with zeolites. In F.R. Ribeiro et al. (eds.), Zeolites : Science and Technology, pp. 317-346, Netherlands, Martinus Nijhoff Publishers, 1984.
48. Ashton, A.G., Batmanian, S., Dwyer, J. Acidity in zeolites. In B. Imelik et al. (eds.), Catalysis by Acids and Bases, pp. 101-109. Amsterdam, Elsevier, 1985.

49. Sano, T., Fujisawa, K., and Higiwara, H. In B. Delmin and G.F. Fromant (eds.),
Stud. Sur. Sci. Catal., 34 : Catalyst Deactivation, pp. 613- 624,
Amsterdam, Elsevier, 1987.
50. Weisz, P.B., Frilette, V.J., Maatman, R.W., and Mower, E.B., J. Catal., 1,
pp. 307-312, 1962.
51. Cricsery, S.M., Zeolites, 4, pp. 202-213, 1984.
52. Anderson, M.W., and Klinowski, J. Nature, 339, pp. 200-203, 1989.
53. Satterfield, C.N., Heterogeneous Catalysis in Practice, New York,
McGraw-Hill Book Co., 1980.
54. Derouane, E. G. New aspects of molecular shape selectivity. In B. Imelik et al.
(eds.), Catalysis by Zeolites, pp. 5-27, Amsterdam, Elsevier, 1980.
55. Burns, K., Collins, M., and Heck, R., Catalytic Control of NO_x Emissions from
Stationary Rich-Burning Natural Gas Engines, ASME83 D6P-12,
1983.
56. Farrauto, R., and Heck, R., Encyclopedia of Chemical Processing and Design,
pp. 218-274, New York, Marcel Dekker.
57. Ronald, M.H., and Farrauto, R.J. Catalytic Air Pollution Control, United State of
America, Van Nostrand Reinhold, 1995.
58. Inui, T., Yamase, O., Fukuda, K., Itoh, A., Tarumoto, J., Morinaga, N.,
Hagiwara, T., and Takegami, Y., Proc. 8th Int. Cong. Catal., Berlin,
pp. 569, Frankfurt-am-Main, Germany, Decema, 1984.
59. T. Inui, H. Matsuda, O. Yamase, H. Nagata, K. Fukada, T. Ukawa,
and A. Miyamoto, J. Catal., 98, pp. 491, 1986.

60. Iwamoto, M., Mizuno, N., Yahiro, H., and Yoshioka, T,
New Developments in Ion Exchange, pp. 407-412, Kodansha,
Tokyo, 1991.
61. Sibilia, J.P., A Guide to Material Characterization and Chemical Analysis,
pp. 102, 116, 258, VCH, New York, 1988.
62. Yogo, K., Tanaka, S., Ihara, M., Hishiki, T.,and Kikuchi, E., Chem. Lett.,
pp. 1025-1028, 1992.
63. Teraoka, Y., Shimane, K., and Yamazoe, Y., Chem. Lett., pp. 2047, 1987.
64. Accomazzo,M. A., and Nobe, K. Catalytic combustion of C₁ to C₃
hydrocarbons, I & EC Process Design and Development, 4,
pp. 425-430, 1965.

APPENDIX

APPENDIX A**SAMPLE OF CALCULATIONS**

M.W. of Si = 28.0855
M.W. of SiO₂ = 60.0843
Weight percent of SiO₂ in Sodium Silicate = 28.5
M.W. of Al = 26.9815
M.W. of AlCl₃ = 133.3405
Weight percent purity of AlCl₃ = 97
M.W. of Cu = 63.54
M.W. of Cu(NO₃)₂·3H₂O = 241.60
M.W. of Cu(CH₃COO)₂ H₂O = 199.65
Weight percent purity of Cu(NO₃)₂·3H₂O = 99.5
Weight percent purity of Cu(CH₃COO)₂ H₂O = 99

A-1 Calculation of Si/Al Atomic Ratio for ZSM-5 and Cu,Al-silicate

The calculation is based on weight of Sodium Silicate (Na₂O·SiO₂·H₂O) in B1 and B2 solutions.

For example, to prepare ZSM-5 at Si/Al atomic ratio of 50.

Using Sodium Silicate 69 g with 45 g of water as B1 solution.

$$\begin{aligned}\text{mole of Si used} &= \frac{\text{wt. \% of SiO}_2}{100} \cdot \frac{\text{wt. of Sod. sil.}}{(\text{M.W. of SiO}_2)} \cdot \frac{(1 \text{ mole})}{(\text{M.W. of Si})} \\ &= 69 \cdot (28.5/100) \cdot (1/60.0843) \\ &= 0.3273\end{aligned}$$

Si/Al atomic ratio = 50

$$\begin{aligned}\text{mole of AlCl}_3 \text{ required} &= 0.3273/50 = 6.5458 \cdot 10^{-3} \text{ mole} \\ \text{amount of AlCl}_3 &= 6.5458 \cdot 10^{-3} \cdot 133.34 (100/97) \\ &= 0.8998 \text{ g}\end{aligned}$$

This is the amount of AlCl₃ used in A1 and A2 solutions

A-2 Calculation of Copper Ion-exchanged ZSM-5 and Metallosilicate

Determine the amount of Cu into catalyst = 2 wt.%

the catalyst use = x g

So that : from the equation

$$\text{Cu}/(x+\text{Cu}) = 2/100$$

$$100 \cdot \text{Cu} = 2 \cdot (x+\text{Cu})$$

$$(100-2) \cdot \text{Cu} = 2 \cdot x$$

thus

$$\text{Cu} = 2 \cdot x / (100-0.2) \text{ g}$$

use $\text{Cu}(\text{CH}_3\text{COO})_2 \cdot \text{H}_2\text{O}$ for copper source

$$\text{weight of } \text{Cu}(\text{CH}_3\text{COO})_2 \cdot \text{H}_2\text{O} = (199.65/63.55) \cdot (100/99) \cdot (2x) / (100-2)$$

A-3 Calculation of Reaction Flow Rate

The catalyst used = 0.50 g

packed catalyst into quartz reactor (diameter = 0.6 cm)

determine the average high of catalyst bed = x cm

So that, volume of catalyst bed = $\pi * (0.3)^2 * x$ ml-catalyst

used GHSV (Gas Hourly Space Velocity) = $4,000 \text{ h}^{-1}$

$$\text{GHSV} = \frac{\text{Volumetric flow rate}}{\text{Volume of Catalyst}} = 4,000 \text{ h}^{-1}$$

$$\begin{aligned}
 \text{Volumetric flow rate} &= 4,000 \cdot \text{Volume of catalyst} \\
 &= 4,000 \cdot \pi (0.3)^2 \cdot x \text{ ml/h} \\
 &= 4,000 \cdot \pi (0.3)^2 \cdot x / 60 \text{ ml/min}
 \end{aligned}$$

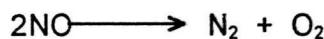
$$\text{at STP : Volumetric flow rate} = \frac{\text{Volume flow rate} \cdot (273.15+t)}{273.15}$$

where : t = room temperature, °C

A-4 Calculation of NO and C₃H₈ conversion

The effluent gas was analyzed by gas chromatography.

-The NO reduction was evaluated in terms of the conversion of NO into N₂.



$$\text{NO Conversion (\%)} = (2[\text{N}_2]_{\text{out}} / [\text{NO}]_{\text{in}}) \times 100$$

$$[\text{NO}]_{\text{in}} = 1000 \text{ ppm}$$

[\text{N}_2]_{\text{out}} : analyzed by gas chromatography

From calibration curve (Figure A1)

Area = area of N₂ peak on GC 8APT

$$\text{Area} \times 10^6 = 0.0276 \times \text{conc. of N}_2 (\%)$$

$$\text{Area} = 2.76 \times 10^4 \times \text{conc. of N}_2 (\%)$$

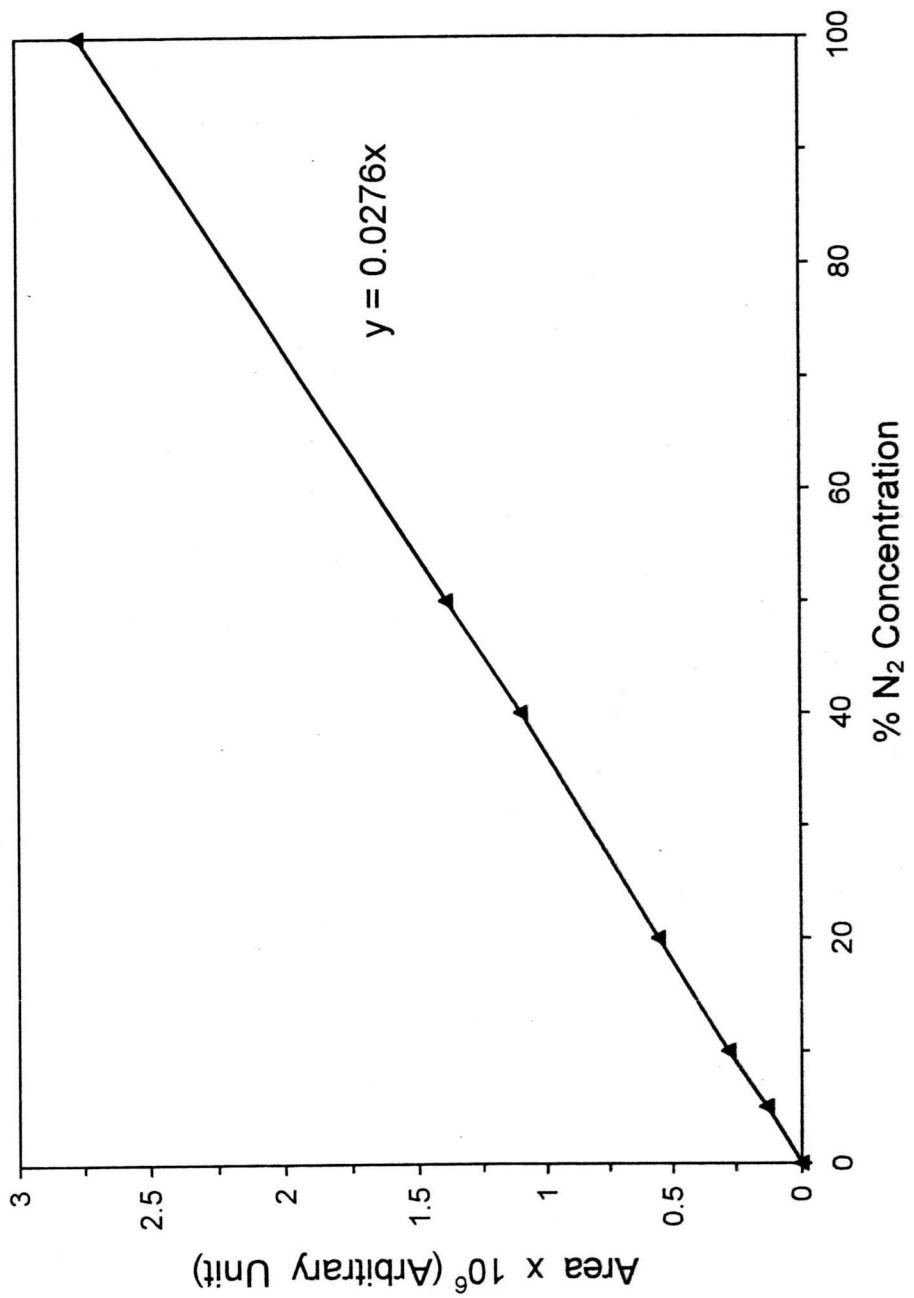


Figure A1 Calibration Curve of N_2 on GC-8APT

$$\text{Area} = 2.76 \times \text{conc of N}_2 \text{ (ppm)} ; (1\% = 10000 \text{ ppm})$$

$$\text{Thus, } [\text{N}_2] = \text{Area} / 2.76$$

- C₃H₈ oxidation was evaluated in terms of the conversion of C₃H₈ into CO and CO₂

$$\text{C}_3\text{H}_8 \text{ Conversion (\%)} = \frac{([\text{C}_3\text{H}_8]_{\text{in}} - [\text{C}_3\text{H}_8]_{\text{out}}) \times 100}{[\text{C}_3\text{H}_8]_{\text{in}}}$$

[C₃H₈]_{in} = conc. of C₃H₈ in reactant gas

(evaluated by areas of C₃H₈ on GC 8AIT)

[C₃H₈]_{out} = conc. of C₃H₈ in product gas

(evaluated by areas of C₃H₈ on GC 8AIT)

APPENDIX B**PHYSICAL AND CHEMICAL PROPERTIES OF NITRIC OXIDE**

Property	Value
mol. wt.	30.1
m.p., °C	-161
b.p., °C	151.18
heat of fusion, kcal/mole	0.550
heat of vaporization, kcal/mole	3.293
heat of formation, kcal/mole	21.50
density [0 °C, 1 atm], g/L	1.2536
sp. gr. [0 °C, 1 atm], (air = 1)	1.269
critical temperature, °C	1.018
critical pressure, atm	64
color	colorless gas, blue liquid and solid

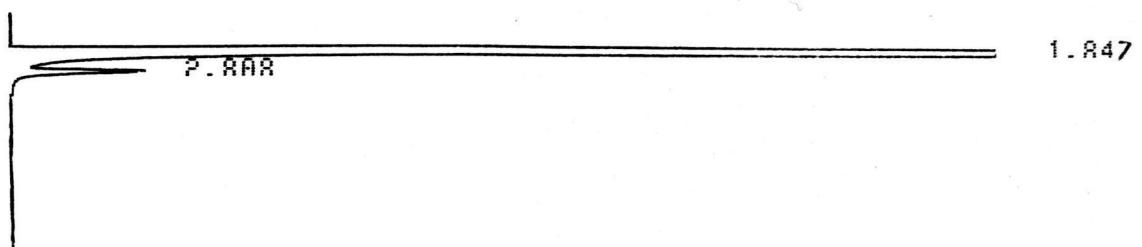
APPENDIX C**AMBIENT AIR QUALITY STANDARD OF THAILAND (1981)**

Pollutants	average value (mg/m ³)				methods of measurement
	1 h	8 h	24 h	1 year	
Carbon Monoxide (CO)	50	20	-	-	Non Dispersive Infrared Detection
Nitrogen Dioxide (NO ₂)	0.32	-	-	-	Gas Phase Chemiluminescence
Sulfur Dioxide (SO ₂)	-	-	0.3	1*	Pararosanniline
Suspended Particulate Matter (SPM)	-	-	0.33	0.1*	Gravimetric
Photochemical Oxidant (O ₃)	0.20	-	-	-	Chemiluminescence
Lead (Pb)	-	-	-	0.01	Wet Ashing

Note : * = Geometric mean

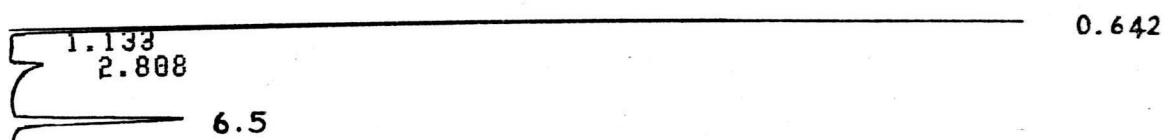
APPENDIX D

SAMPLE OF CHROMATOGRAMS



COLUMN MS-5A

PKNO	TIME	AREA	CONC	NAME
1	1.847	233466	99.4342	O ₂
2	2.808	1329	0.5658	N ₂



COLUMN PORAPAK-Q

PKNO	TIME	AREA	CONC	NAME
1	0.642	69558	93.2220	AIR
2	2.808	977	1.3095	H ₂ O
3	6.500	4080	5.4684	C ₃ H ₈

VITA

Miss Woraratana Pattaraprakorn was born in Bangkok, Thailand, on August 30, 1972. She received her Bachelor Degree of Science from the Department of Chemical Technology, Faculty of Science, Chulalongkorn University in 1994.